

Coherent Optics and Localized Light

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Abstract

Interplay between the effects of coherent radiation and localization of light is analysed. A system of two-level atoms is placed in a medium interacting with electromagnetic field. The matter-light interaction can result in the appearance of a band gap in the spectrum of polariton states. If an atom with a resonance frequency inside the gap is incorporated into such a medium, the atomic spontaneous emission is suppressed, which is termed the localization of light. However, a system of resonance atoms inside the gap can radiate due to their coherent interactions. The peculiarity of the coherent radiation by a system of atoms, under the localization of light for a single atom, is studied.

1 Localization of Light

In some cases a medium may possess an electromagnetic band gap where a severe depressions of the photon density of states occurs. One such case is rather well known – This is the appearance of the polariton band gap due to the interaction of light with a dense medium [1-3]. Another possibility has been recently found when a spectral photon gap develops because of periodicity of dielectric structures [4,5]. The latter are called photonic band-gap materials. In such dielectric superlattices, strong localization of photons happens [4-6]. The same effect of light localization arises if a resonance atom, with a frequency inside the polariton band gap, is doped into a dispersive medium [7-9]. If the atomic resonance frequency lies near the gap, a polariton–atom bound state appears with an eigenfrequency lying within the gap. The appearance of this bound state results in a significant suppression of spontaneous emission, that is, in localization of light. This means that an atom, in the stationary state, has a finite probability to be in the excited state, provided that it was excited at the initial moment of time.

To explain in simple parlance what does mean the localization of light, let us consider a two-level atom whose population difference is described by an operator $\sigma^z(t)$. It is convenient to introduce the excitation function

$$\eta(t) \equiv \frac{1}{2} (1 + s(t)), \quad s(t) \equiv \langle \sigma^z(t) \rangle,$$

in which the angle brackets $\langle \dots \rangle$ imply a quantum-mechanical averaging. The atom is excited when the population difference $s = 1$, i.e., the excitation function $\eta = 1$. When the atom is not excited, then $s = -1$ and $\eta = 0$. There exists a finite probability to find the atom excited, in the stationary state, if and only if

$$\lim_{t \rightarrow \infty} \eta(t) \neq 0, \quad \lim_{t \rightarrow \infty} s(t) = \zeta \neq -1.$$

This is exactly what one means under the *localization of light*. The reason for this localization is the suppression of spontaneous emission, if the atom was initially excited. If the suppression is absolute, then, starting with $s(0) = 1$, one comes to $\zeta = 1$, that is, the atom does not become deexcited, keeping an absorbed photon forever. Vice versa, if an atom, with a frequency inside the polariton band gap, was not excited at the initial time $t = 0$, that is,

$s(0) = -1$, then it should remain not excited in the stationary state, as $t \rightarrow \infty$, since there are no photons in the gap, which could excite the atom. The corresponding dynamics of the population difference can be described by the equation

$$\frac{ds}{dt} = -\gamma_1(s - \zeta), \quad \zeta \approx s(0),$$

whose evident solution $s(t) = \zeta \approx s(0)$ demonstrates the suppression of spontaneous emission.

The situation becomes more complicated when a collection of identical impurity atoms, with a transition frequency in the polariton band gap, is incorporated into the medium. If the spacing between the admixture atoms is much smaller than the transition wavelength, then the electromagnetic coupling of atoms leads to the formation of a photonic impurity band within the polarization band gap [10]. If the density of the admixture is high, the polariton gap can be destroyed at all. Electromagnetic field can propagate in the impurity band formed by collective interactions of atoms, and coherent radiation becomes possible. If the spacing between the admixture atoms would be much larger than the transition wavelength, then the propagation band could not be formed and the atomic radiation would be prohibited. In this way, only coherent interactions can overcome the suppression of emission caused by the localization of light.

The time evolution of spontaneous emission near the edge of a photonic band gap has been considered [11,12] for a simple concentrated Dicke model, where the radiation wavelength is assumed to be much larger than not only the interatomic spacing but the whole system. This model, evidently, is equivalent to a single-atom model with atomic variables factored by the number of atoms.

The aim of the present communication is to suggest a more realistic, though yet solvable, approach to describing coherent emission of admixture atoms placed in a medium with localizing light. Clearly, a more realistic approach is, at the same time, more and even much more complicated. Therefore, it would be impossible in the frame of this communication to expound it in whole. The main attention here is paid to the the formulation of the problem, with a brief survey of physical picture, and some new results are announced.

2 Formulation of Problem

Consider a system of N resonance two-level radiators enumerated by the index $i = 1, 2, \dots, N$. These can be atoms, molecules, nuclei, or quantum dots. For short, let us call them atoms. Their Hamiltonian is

$$\hat{H}_a = \frac{1}{2} \sum_{i=1}^N \omega_0 (1 + \sigma_i^z), \quad (1)$$

where ω_0 is a transition frequency ($\hbar \equiv 1$) and σ_i^z is a population difference operator. The electromagnetic field Hamiltonian has the general form

$$\hat{H}_f = \frac{1}{8\pi} \int \left[\vec{E}^2(\vec{r}) + \vec{H}^2(\vec{r}) \right] d\vec{r}, \quad (2)$$

with the electric field \vec{E} and magnetic field $\vec{H} \equiv \vec{\nabla} \times \vec{A}$, where \vec{A} is the vector-potential satisfying the Coulomb gauge condition $\vec{\nabla} \cdot \vec{A} = 0$. The interaction between the atoms and field, in the dipole approximation, is given by

$$\hat{H}_{af} = -\frac{1}{c} \sum_{i=1}^N \vec{J}_a(\vec{r}_i) \vec{A}(\vec{r}_i), \quad (3)$$

with the transition current

$$\vec{J}_a(\vec{r}_i) = i\omega_0 \left(\sigma_i^+ \vec{d}^* - \sigma_i^- \vec{d} \right), \quad (4)$$

in which σ_i^\pm is a raising or lowering operator, respectively; and \vec{d} , a transition dipole. As usual [13], the relativistic term \vec{A}^2/c^2 is neglected. The system of radiators defined by Eqs.(1) to (4) forms the basis for the standard consideration of collective processes in the emission of light [14,15].

The case we are considering here is aggravated by the fact that the resonance atoms are not in empty space but are inserted as admixtures into a medium. The latter can be modeled in different ways, with the main requirement that a band gap should appear resulting in the localization of light for a single atom. Photonic band-gap materials can be described by periodic superstructures of scatterers [4,5]. A frequency gap for propagating electromagnetic modes exists also in many natural dielectrics and semiconductors

[1,2]. For instance, the polariton effect is well developed in such semiconductors as $CuCl$, $CuBr$, CdS , $CdSe$, $ZnSe$, $GaAs$, $GaSb$, $InAs$, $AlAs$, SiC , and in some semiconductor microstructures including quantum dots, wells, and wires [16]. A frequency gap for light propagation inside dense media is known to appear when the latter contain excitations being in resonance with the frequency of light [1,2]. For example, the matter could be presented as an ensemble of two-level radiators. Then, these radiators together with the admixture atoms would form a kind of a resonance two-component system [17-19]. The polariton gap in dispersive dense media arises because of the interaction of light with some gapful elementary excitations, like excitons or optical phonons [1,2]. Hence, a medium can be modeled by an ensemble of oscillators, possessing an optical branch, which represent optical-type collective excitations of the medium. A Hamiltonian of such collective excitations has, in general, the form

$$\hat{H}_m = \sum_{j=1}^{N'} \frac{\vec{p}_j^2}{2m} + \frac{1}{2} \sum_{ij}^{N'} \sum_{\alpha\beta}^3 D_{ij}^{\alpha\beta} u_i^\alpha u_j^\beta, \quad (5)$$

where N' is the number of lattice sites; \vec{p}_i and \vec{u}_i are momentum and displacement operators, respectively; $D_{ij}^{\alpha\beta}$ is a dynamical matrix. The interaction of the matter excitations with electromagnetic field is described by the term

$$\hat{H}_{mf} = -\frac{1}{c} \sum_{j=1}^{N'} \vec{J}_m(\vec{r}_j) \vec{A}(\vec{r}_j), \quad (6)$$

in which

$$\vec{J}_m(\vec{r}_j) = \frac{e}{m} \vec{p}_j \quad (7)$$

is a local current at the point \vec{r}_j of the medium.

In this way, the total Hamiltonian of the considered system is

$$\hat{H} = \hat{H}_a + \hat{H}_f + \hat{H}_{af} + \hat{H}_m + \hat{H}_{mf}, \quad (8)$$

consisting of the atom Hamiltonian (1), field term (2), atom-field interaction (3), medium Hamiltonian (5), and of the medium-field interaction (6).

The stationary states of the system with Hamiltonian (8) can be studied as follows. One expands the vector potential

$$\vec{A} = \sum_{k\nu} \left(\frac{2\pi c}{kV} \right)^{1/2} \left(a_{k\nu} \vec{e}_{k\nu} e^{i\vec{k}\vec{r}} + a_{k\nu}^\dagger \vec{e}_{k\nu}^* e^{-i\vec{k}\vec{r}} \right) \quad (9)$$

in plane waves, with $\vec{e}_{k\nu}$ being a polarization vector; $k \equiv |\vec{k}|$; \vec{k} , a wave vector; $\nu = 1, 2$; V , volume; and $a_{k\nu}$ being a photon operator indexed by the wave vector \vec{k} and the polarization–branch number ν . The displacement and momentum operators of the medium are also expanded in plane waves:

$$\begin{aligned}\vec{u}_j &= \sum_{ks} (2mN'\omega_{ks})^{-1/2} (b_{ks} + b_{-ks}^\dagger) \vec{e}_{ks} e^{i\vec{k}\cdot\vec{r}_j}, \\ \vec{p}_j &= -i \sum_{ks} \left(\frac{m\omega_{ks}}{2N'} \right)^{1/2} (b_{ks} - b_{-ks}^\dagger) \vec{e}_{ks} e^{i\vec{k}\cdot\vec{r}_j},\end{aligned}\quad (10)$$

where \vec{e}_{ks} is a corresponding polarization vector; $s = 1, 2, 3$; and ω_{ks} is the spectrum of collective excitations defined by the eigenvalue problem

$$\frac{1}{mN'} \sum_{ij} \sum_{\beta=1}^{N'} D_{ij}^{\alpha\beta} e^{i\vec{k}\cdot\vec{r}_{ij}} e_{ks}^\beta = \omega_{ks}^2 e_{ks}^\alpha, \quad (11)$$

with $\vec{r}_{ij} = \vec{r}_i - \vec{r}_j$. The frequency and polarization vectors are assumed to be even functions of the wave vector, $\omega_{ks} = \omega_{-ks}$, $\vec{e}_{ks} = \vec{e}_{-ks}$. The destruction, b_{ks} , and creation, b_{ks}^\dagger , operators of collective oscillations satisfy the Bose commutation relations. With expansion (9), the field Hamiltonian (2) takes the known simple form

$$\hat{H}_f = \sum_{k\nu} ck \left(a_{k\nu}^\dagger a_{k\nu} + \frac{1}{2} \right). \quad (12)$$

And the matter Hamiltonian (5), by means of (10), becomes

$$\hat{H}_m = \sum_{ks} \omega_{ks} \left(b_{ks}^\dagger b_{ks} + \frac{1}{2} \right). \quad (13)$$

Recall that an optical-type spectrum ω_{ks} is to be assumed in (13). Introducing polariton operators that are linear combinations of the photon operators $a_{k\nu}$ and of the boson operators $b_{k\nu}$, one can, in some cases [1,2], diagonalize the sum of the Hamiltonians $\hat{H}_f + \hat{H}_m + \hat{H}_{mf}$, obtaining a diagonal polariton Hamiltonian. For example, a detailed description of this procedure of diagonalization can be found in Ref.[20], where a uniform and isotropic model is considered and several simplifications, in line with the Heitler–London and

resonance approximation, are involved. In these approximations, one neglects the counter-rotating terms and two-boson transitions, whose influence, similarly to two-atom transitions [21], can become important only far from the resonance.

When there is only one admixture atom in the medium, that is $N = 1$, then the stationary states of Hamiltonian (8) can be found [9,20] resorting to the uniform, isotropic, and resonance approximations. If the atomic resonance frequency lies near the gap, there appears a polariton-atom bound state with an eigenfrequency lying within the gap. This means that light is localized at the atom. As a result of the appearance of this localized bound state, a significant suppression of spontaneous emission occurs [9]. The behavior of many-polariton states is a little more diverse. Those states containing an even number of polaritons correspond to solitons that can propagate within the gap, while the states with an odd number of polaritons represent solitons that are pinned to the atom forming a many-polariton bound state. The latter state, similarly to the single-polariton bound state, also depicts the localization of light [20].

When two identical two-level atoms are placed in a frequency dispersive medium whose polariton spectrum has a gap, then the polariton-atom bound state lying within the polaritonic gap splits into a doublet due to an effective atom-atom interaction [22]. The spontaneous emission can exist only if the resonance frequency of these two atoms lies in the polariton continuous spectrum. And if the resonance frequency lies within the gap, then two discrete modes represent a doublet of bound polariton-atom states, for which spontaneous emission is practically completely suppressed [22,23].

A qualitatively different situation develops when many resonance admixture atoms are placed in the medium. Stationary states for this case have been studied for a one-dimensional atomic chain incorporated in a uniform and isotropic system [22,24]. The nearest-neighbor approximation has been used. In the case of spatially correlated atoms, with a frequency inside the gap, a polariton-impurity band is formed within the polaritonic gap. Then polaritons can propagate in this impurity band and the atomic chain provides a waveguide for the radiation field. Even in the nearest-neighbor approximation, when, for a chain, the interaction of only three atoms is effectively taken into account, the width of the impurity band, normalized with respect to the polariton gap, is 0.14 [24]. Since this width is, roughly speaking, proportional to $N - 1$, the collective interaction of about 10 atoms should fill

the whole polariton gap.

The energy–momentum representation employed for studying stationary states is not convenient for considering space–time dynamics of collective processes. For the latter purpose, to our mind, it is more appropriate to remain in real space and time. One may write the Heisenberg evolution equations for the operators of the problem. Then, formally solving the Maxwell equations, one may exclude the field variables (see details in [25]). After that, one comes to the equations for the atomic variables,

$$\begin{aligned} \frac{d\sigma_i^-}{dt} = & -(i\omega_0 + \gamma_2)\sigma_i^- + \sigma_i^z \vec{d}^* \cdot \vec{D}_i + \\ & + ik_0^2 \sigma_i^z \vec{d} \cdot \sum_{j(\neq i)}^N \frac{1}{r_{ij}} \left[\sigma_j^+ \left(t - \frac{r_{ij}}{c} \right) \vec{d}^* - \sigma_j^- \left(t - \frac{r_{ij}}{c} \right) \vec{d} \right], \end{aligned} \quad (14)$$

$$\begin{aligned} \frac{d\sigma_i^z}{dt} = & -\gamma_1 (\sigma_i^z - \zeta) - 2 \left(\sigma_i^+ \vec{d}^* + \sigma_i^- \vec{d} \right) \cdot \vec{D}_i - \\ & - 2ik_0^2 \left(\sigma_i^+ \vec{d}^* + \sigma_i^- \vec{d} \right) \sum_{j(\neq i)}^N \frac{1}{r_{ij}} \left[\sigma_j^+ \left(t - \frac{r_{ij}}{c} \right) \vec{d}^* - \sigma_j^- \left(t - \frac{r_{ij}}{c} \right) \vec{d} \right], \end{aligned} \quad (15)$$

where γ_1 and γ_2 are the longitudinal and transverse relaxation parameters, respectively; $k_0 \equiv \omega_0/c$; $r_{ij} \equiv |\vec{r}_{ij}|$; and

$$\vec{D}_i(t) = \frac{k_0}{c} \sum_{j(\neq i)}^{N'} \frac{1}{r_{ij}} \vec{J}_m \left(\vec{r}_j, t - \frac{r_{ij}}{c} \right)$$

is an electric field produced by the medium. Assuming that the single–atom stationary state corresponds to localized light, we put $\zeta = \langle \sigma_i^z(0) \rangle$, where $\langle \dots \rangle$ means the statistical averaging over an initial state. Equations (14) and (15) are complemented by initial conditions

$$u_0 = \langle \sigma_i^-(0) \rangle, \quad s_0 = \langle \sigma_i^z(0) \rangle.$$

3 New Results

The notion of light localization is relatively recent. This is why the main part of this communication has been devoted to the description of the related physical picture and to the formulation of the basic equations that

would permit one to consider the time development of collective phenomena for a system of admixture atoms in a medium with localized light. The limited frames of this communication do not allow to expound in detail the way of solving the basic equations (14) and (15) and the analysis of the corresponding solutions. This will be done in a separate publication. Here, we only can briefly delineate the scheme of solving Eqs.(14) and (15) and present some fresh results.

From the operator equations (14) and (15), we pass to the equations for the related statistical averages. The pair correlation functions are decoupled in the semiclassical approximation. The retardation is treated in the quasirelativistic approximation as in Refs. [26-28]. The system of nonlinear differential equations is solved by means of the scale separation approach [29,30]. The resulting physical picture depends on the values of the following parameters: initial conditions u_0 and s_0 ; the coupling parameter of coherent atomic interactions,

$$g = \frac{k_0^3 d_0^2}{\gamma_2} \sum_{j(\neq i)}^N \frac{\sin(k_0 r_{ij})}{k_0 r_{ij}}; \quad (16)$$

the effective parameter of coupling between the admixture atoms and matter,

$$\alpha = \langle\langle \left| e^{-\Gamma t} \int_0^t e^{(i\Omega+\Gamma)\tau} \vec{d}^* \cdot \vec{D}(\tau) d\tau \right|^2 \rangle\rangle, \quad (17)$$

where the double brackets $\langle\langle \dots \rangle\rangle$ imply the statistical and time averaging, and

$$\Omega = \omega_0 + \Delta_L s, \quad \Gamma = \gamma_2(1 - gs) \quad (18)$$

are an effective frequency and attenuation, with

$$\Delta_L = k_0^3 d_0^2 \sum_{j(\neq i)}^N \frac{\cos(k_0 r_{ij})}{k_0 r_{ij}}, \quad s = \langle \sigma_i^z \rangle;$$

and the critical atom-matter coupling parameter

$$\alpha_c = \frac{(1 - gs_0)^2 + 4g^2|u_0|^2}{4g^2s_0^2}. \quad (19)$$

The overall physical picture for the case $g \gg 1$ and $\alpha \ll \alpha_c$ is as follows. If the admixture atoms at the initial time $t = 0$ are excited, then after a delay

time $t_0 \ll T_1$, defined by the values of the above parameters, a coherent burst occurs. Then, a series of coherent bursts follows, separated from each other by the periods of practically no radiations. This series of bursts lasts for the time of several T_1 . Finally, dynamics tends to a stationary state with the excitation function

$$\lim_{t \rightarrow \infty} \eta(t) = \frac{1}{2} \left(1 + \frac{1}{g} \right). \quad (20)$$

Remembering the definition in Section 1, we see that the limit (20) exhibits a partial localization of light.

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References

- [1] Davydov, A.S., 1971, *Theory of Molecular Excitons* (New York: Plenum).
- [2] Agranovich, V.M. and Ginzburg, V.L., 1984, *Crystal Optics with Spatial Dispersion and Excitons* (Berlin: Springer)
- [3] Zheleznyakov, V.V., Kocharovskiy, V.V., and Kocharovskiy, Vl.V., 1989, *Phys. Usp.*, **32**, 835.
- [4] Yablonovitch, E., 1987, *Phys. Rev. Lett.*, **58**, 2059.
- [5] John, S., 1987, *Phys. Rev. Lett.*, **58**, 2486.
- [6] John, S., 1984, *Phys. Rev. Lett.*, **53**, 2169.
- [7] Rupasov, V.I. and Singh, M., 1996, *J. Phys. A*, **29**, L 205.
- [8] Rupasov, V.I. and Singh, M., 1996, *Phys. Rev. Lett.*, **77**, 338.
- [9] Rupasov, V.I. and Singh, M., 1996, *Phys. Lett. A*, **222**, 258.
- [10] John, S. and Wang, J., 1991, *Phys. Rev. B*, **43**, 12772.
- [11] John, S. and Quang, T., 1994, *Phys. Rev. A*, **50**, 1764.
- [12] John, S. and Quang, T., 1995, *Phys. Rev. Lett.*, **74**, 4319.
- [13] Allen, L. and Eberly, J.H., 1975, *Optical Resonance and Two-Level Atoms* (New York: Wiley).
- [14] Andreev, A.V., Emelyanov, V.I., and Ilinski, Y.A., 1993, *Cooperative Effects in Optics* (Bristol: Inst. of Physics).
- [15] Benedict, M.G., Ermolaev, A.M., Malyshev, V.A., Sokolov, I.V., and Trifonov, E.D., 1996, *Superradiance–Multiatomic Coherent Emission* (Bristol: Inst. of Physics).
- [16] Ivanov, A.L., Haug, H., and Keldysh, L.V., 1998, *Phys. Rep.*, **296**, 237.

- [17] Bogolubov, N.N., Turaev, M.T., Shumovsky, A.S., and Yukalov, V.I., 1986, *JINR Rapid Commun.*, **14**, 33.
- [18] Andreev, A.V. and Polevoy, P.V., 1994, *Quantum Opt.*, **6**, 57.
- [19] Andreev, A.V. and Polevoy, P.V., 1996, *Quantum Electron.*, **26**, 724.
- [20] Rupasov, V.I. and Singh, M., 1996, *Phys. Rev. A*, **54**, 3614.
- [21] Antipin, E.L. and Gadomsky, O.N., 1996, *Teor. Mat. Fiz.*, **106**, 145.
- [22] Rupasov, V.I. and Singh, M., 1997, *Phys. Rev. A*, **56**, 898.
- [23] Singh, M.R. and Lau, W., 1997, *Phys. Lett. A*, **231**, 115.
- [24] Singh, M.R. and Lau, W., 1997, *Phys. Status Solidi B*, **203**, 401.
- [25] Yukalov, V.I., 1991, *Laser Phys.*, **1**, 85.
- [26] Emelyanov, V.I. and Yukalov, V.I., 1986, *Opt. Spectrosc.*, **60**, 385.
- [27] Yukalov, V.I., 1988, *J. Mod. Opt.*, **35**, 35.
- [28] Yukalov, V.I., 1990, *J. Mod. Opt.*, **37**, 1361.
- [29] Yukalov, V.I., 1995, *Phys. Rev. Lett.*, **75**, 3000.
- [30] Yukalov, V.I., 1996, *Phys. Rev. B*, **53**, 9232.